GDP-0392 March 10, 1988

RADIONUCLIDES IN THE EQUIPMENT OF THE OAK RIDGE GASEOUS DIFFUSION PLANT*

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During the more than 30 years of operations at the Oak Ridge Gaseous Diffusion Plant many thousands of tons of uranium have been processed through the diffusion cascade. While the vast majority of this uranium has consisted of naturally occurring material, some has been obtained from spent reactor fuel.

The processing of uranium from spent reactor material has been intermittent. The first campaign lasted from 1952 through 1964, the second from 1969 through 1974, and the third from 1976 through 1977. Virtually all of this uranium came from military plutonium production reactors at Hanford and Savannah River and very little was obtained from commercial power reactors.

Unlike natural uranium the material recovered from reactor fuels contains fission products and transuranic elements formed during irradiation of the uranium-containing fuels. Most of these materials were removed from the uranium during the recovery process, but traces of these substances did carry through into the UF_6 and were introduced into the cascade.

SLIDE 1

The principal isotopes of concern are technetium-99, plutonium-239, and ›tunium-237. Two additional isotopes which seem to offer potentials for ..me very serious problems for the recycle of reactor fuels are ruthenium-106 and antimony-125. All of these isotopes get through the uranium-recovery operation in sufficient quantities to be troublesome and they also form volatile fluorides so they carry through into the UF_6.

Ruthenium-106 and Antimony-125

I would like to deal with the ruthenium-106 and the antimony-125 first because we can get them out of the way rather easily and then go on the other isotopes which are more troublesome.

The volatile fluorides of ruthenium and antimony readily convert to non-volatile compounds on contact with the surfaces of the cascade equipment. This characteristic would cause the materials to deposit in the first stage or two as they are introduced into the cascade. The daughter products of these isotopes are potent gamma radiation emitters, and any such deposits could produce gamma radiation fields sufficiently high to pose radiation protection difficulties. In addition, at radiation levels far below any real health physics concern, such radiation fields could make it impossible to find deposits of uranium materials inside the cascade equipment by means of gamma radiation measurements at the exterior surfaces of the equipment.

For many years the cascade equipment was surveyed with sensitive gamma ›diation detectors to locate any deposits of uranium materials in the ..quipment. The detection of a deposit depended on the measurement of the

Paper was presented by J. C. Bailey at a D&D Briefing held at the ORGDP for DOE Oak Ridge Operations and HQs personnel, on March 10, 1988.

*Based on work performed at the Oak Ridge Gasous Diffusion Plant managed by Martin Marietta Energy Systems, Inc., for the U.S. Department of Energy under

OR0030895

SELECTED PROPERTIES OF IMPORTANT RADIONUCLIDE IMPURITIES IN REACTOR RETURN UF_6*

Nuclide	Radioactive decay mode and energy, Mev.**	Half-life (yr)	Unstable decay daughter products
Tc-99	Negative beta, 0.292	2.12×10^5	None
Ru-106	Negative beta, 0.0394	1.0	Rh-106, 3.54 Mev; decays with a half life of 30s to stable Pd-106
Sb-125	Negative beta, 0.764	2.7	Te-125m; decays with a half life of 58 days to stable Te-125
Np-237	Alpha, 4.956	2.14×10^6	Pa-233. Initial product of disintegration series.
Pu-239	Alpha, 5.243	2.44×10^4	U-235. Initial product of disintegration series

195 Mev gamma ray associated with the alpha radiation decay of U-235. The els of radiation being measured amounted to a few tens of microroentgens per hour, and any background radiation on the order of 0.5 mr/hr would be sufficient to make detection of this gamma radiation impossible, even with high resolution gamma spectroscopy techniques. It was therefore a matter of great concern that the introduction of such elements be very closely controlled and limited.

Technetium-99

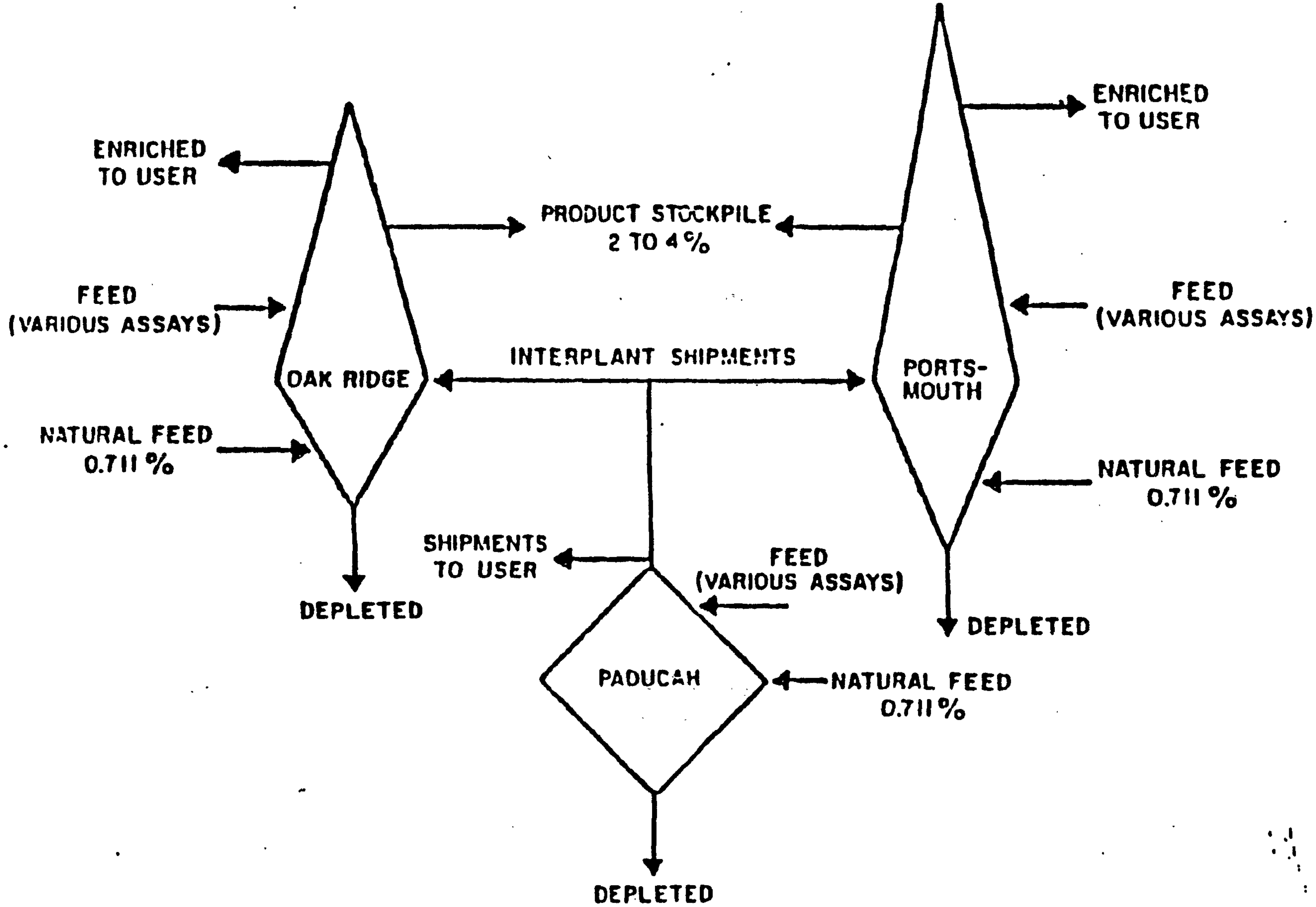
Most of the information I am going to present about technetium comes from a draft report prepared under the direction of Mr. R. W. Levin in 1979. Mr. Levin was the director of the ORGDP Technical Services Division, but both the Portsmouth Plant and the Paducah Plant were heavily involved in bringing this information together. Unfortunately, the report was never issued, but it is probably the best source of information on the subject available.

Technetium is very close to uranium in its chemical nature and forms compounds which carry through the chemical separation processes to a nificant degree. A number of its compounds, TcF_6 for example, are seous under cascade conditions, and being of lower molecular weight than UF_6, diffused preferentially toward the enriched part of the cascade. This transport of Tc did not take place rapidly, however; the Tc compounds are deposited on internal surfaces of the cascade equipment and then, over a period of several years, they migrate toward the enriched portion of the cascade. In the case of Paducah Tc was found in the product about 2 years after the feeding of reactor return materials started. At Portsmouth the period was 20 years.

Most of the reactor return materials, and therefore most of the technetium, was fed into the Paducah Diffusion Plant. Information regarding the fate of technetium, plutonium, and neptunium at the Paducah Plant was more fully developed than is the case at either of the other diffusion plants. Much of the technetium initially fed at the Paducah Plant was later shipped to the other plants in Paducah product material. In order to track the flows of technetium among the diffusion plants let's look at a diagram of the interplant flows of UF_6.

SLIDE 2

The thing I want to call your attention to is that UF_6 fed to the Paducah Plant, after partial enrichment, is shipped to both the Oak Ridge and Portsmouth Plants as feed materials. Product at Paducah is withdrawn from the ne cell in which technetium tends to accumulate, so much of the Tc which ssed through the Paducah Plant was shipped to the Oak Ridge and Portsmouth sites. The quantities of Tc received at each of the sites are listed in the next slide.



DIFFUSION PLANT COMPLEX

SLIDE 3

TECHNETIUM RECEIVED

Source of Technetium	Amount of Technetium Received at Respective Plant, kg		
	GAT	ORGDP	PGDP
Hanford and Savannah River	<0.5	86	606
ORGDP (nonenriched UF_6)	0	-	64
PGDP (enriched UF_6)	79	117	-
PGDP (nonenriched UF_6)	6	4	-
Commercial Reactors	0	<0.04	0
Total Receipts*	85	207	670

* This total includes the transfers between plants. Actual quantity of technetium received is reflected by the top line, i.e., 692 kg.

Notice that Oak Ridge received 86 kg of Tc from Hanford and Savannah River while Paducah received 606 kg from these plants. Oak Ridge also received 117 kg in Paducah product; part of this is from the 606 kg that Paducah got from the reactor sites. Also notice the very small contribution from the commercial reactors, <0.04 kg.

The best estimates regarding the distribution of Tc in the Paducah Plant are that approximately 70% of the Tc was absorbed on the equipment, with most of the remaining 30% being distributed between the product and tails stream. In view of the preferential movement of the Tc compounds toward the enriched portion of the cascade it is reasonable to assume that most of this last 30% left the plant in the product stream. It is also reasonable to assume a similar distribution of the Tc introduced into the ORGDP cascade equipment with the exception of the amount leaving in the product stream. As noted previously Paducah product is withdrawn from the cell in which Tc would tend to concentrate, and much of the Tc therefore passes out with the product.

At the Oak Ridge Plant the functioning of the purge system changed this situation drastically. The purge cascade was in the K-311-1 and K-310-3 units until 1977 when the K-402-9, K-402-8 purge system started operations. Product was withdrawn from the K-29 Plant, well below this purge cascade. Thus, very little Tc left ORGDP in the product.

K-402-8 operated as a side purge unit to remove gasses of intermediate molecular weight, such as gasses due to inleakages from the coolant system. These intermediate weight gasses were moved out the top of the K-402-8 side purge system through sodium-fluoride traps and were vented. The Tc compounds, being of intermediate molecular weight also moved out by this route but were absorbed in the trap along with UF_6. The absorption mechanism is reversible at elevated temperatures, and the trap was subsequently heated and the UF_6 and technetium compounds were returned to the main purge system. Some of the technetium was removed from the purge system by elevating the temperature of the equipment and trapping the technetium which was mobilized by the higher temperatures. Approximately 100 pounds of technetium and uranium compounds were trapped in this way, but the fraction of this material which is technetium is not known.

If we gauge the fate of technetium in the ORGDP system on the basis of Paducah's experience, it would appear that approximately 70% of the 207 kg of technetium received, or about 145 kg, remained in the cascade equipment. Most of the remaining 62 kg might be considered to remain in the purge cascade equipment, except for the unknown amount of material trapped. We might expect about 32 kg to be in the K-311-1, K-310-3 system, the remaining being in the K-402-8, K-402-9 system or in the trapped material.

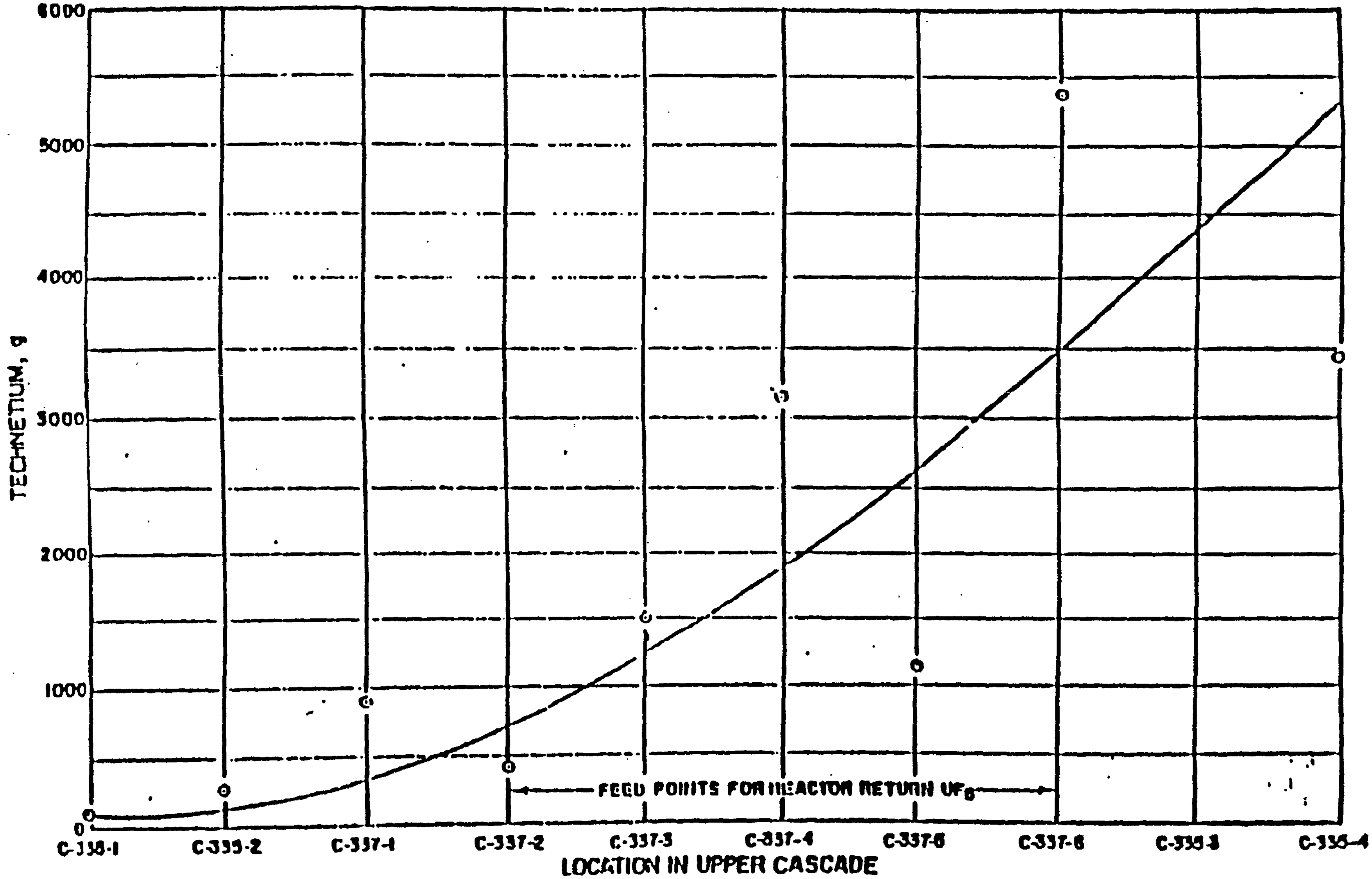
The pattern of the technetium deposited in the Paducah cascade is shown the next slide.

SLIDE 4

Taking this as the general pattern which would have occurred at ORGDP it is obvious that high concentrations of Tc would be expected in the K-29 Plant in the units constituting the top of the enrichment cascade. These units are K-502-1 and cells 1, 3, and 5 in K-502-2. K-502-3 and the other cells of K-502-2 were shut down in 1968. From 1964, when the K-27 Plant was shut down, until 1968 all of the K-29 Plant represented to top of the enrichment cascade. The presence of significant quantities of technetium in the K-29 Plant was clearly evident from radiation monitoring reports from this area.

Technetium compounds left in the operating cascade equipment in the K-31 and K-33 parts of the plant were were nearly all removed during the cascade improvement program. These compounds would have been removed from the equipment during the decontamination cycle and would have been discarded through the waste streams.

There are no data available to clearly identify how much technetium is currently in the K-25 and K-27 equipment other than the purge systems. However, the monitoring of equipment removed from these units, carried out as part of the routing health physics surveillance program, did not reveal any technetium problems. It is therefore considered that no significant technetium accumulations exist in these areas.



DEPOSITION OF TECHNETIUM IN THE UPPER CASCADE

Health Physics Aspects of Technetium

Technetium emits beta radiation with a maximum energy of 0.29 Mev. This radiation is completely stopped by a thickness of material equivalent to heavy neoprene gloves or 2 feet of air. This radiation can produce high skin radiation exposures if the skin is exposed directly to the technetium. Technetium compounds may also become airborne and cause internal radiation exposure if they are inhaled. Only in the purge-cascade equipment, and to a lesser degree in the K-29 equipment, does technetium present significant radiation exposure potentials.

In equipment removal or maintenance operations relatively simple safety measures permit the activities to be conducted without personnel radiation exposure. Prescribed safety measures for opening or removing equipment of the UF_6 system in the purge cascade include:

1. Company shoes and clothing.
2. Full face respirator with a combination acid-gas/particulate filter.
3. Heavy, elbow-length neoprene gloves.
4. Full disposable suit and hood over Company clothing and shoes.
5. Training in appropriate removal of protective items.
6. Personnel monitoring upon completion of the job and after removal of disposable suit, respirator, and gloves.
7. There are also requirements for isolating areas of work and for the entry of personnel into the area other than those directly involved in the work.

Neptunium and Plutonium

Documented information regarding the disposition of technetium among the plants is probably the most reliable index we have for evaluating the distribution of plutonium and neptunium also. The next slide lists the amounts of Tc, Np, and Pu received by Paducah and the amounts actually fed to the cascade. In the case of technetium reasons for the difference between the amount received and the amount fed is not clear. In the cases of neptunium and plutonium it is known that large fractions of these materials remain the the feed cylinders and are not transferred to the cascade. If we take the value of 86 kg as the amount of technetium received from Hanford and Savannah River and fed at ORGDP, and 529 kg as the quantity received and fed at Paducah, we can then calculate proportional amounts of plutonium and neptunium which would have been received at Oak Ridge. Values so derived are also shown on this slide.

SLIDE 5

QUANTITIES OF TECHNETIUM, NEPTUNIUM,
AND PLUTONIUM FED AT THE PADUCAH DIFFUSION PLANT

Material	Quantity Received	Quantity Fed to Cascade
Tc-99	670 kg	529 kg
Np-237	13.5 kg	3.3 kg
Pu-239	271 g	0.4 g

QUANTITIES OF TECHNETIUM, NEPTUNIUM,
AND PLUTONIUM FED AT THE OAK RIDGE DIFFUSION PLANT

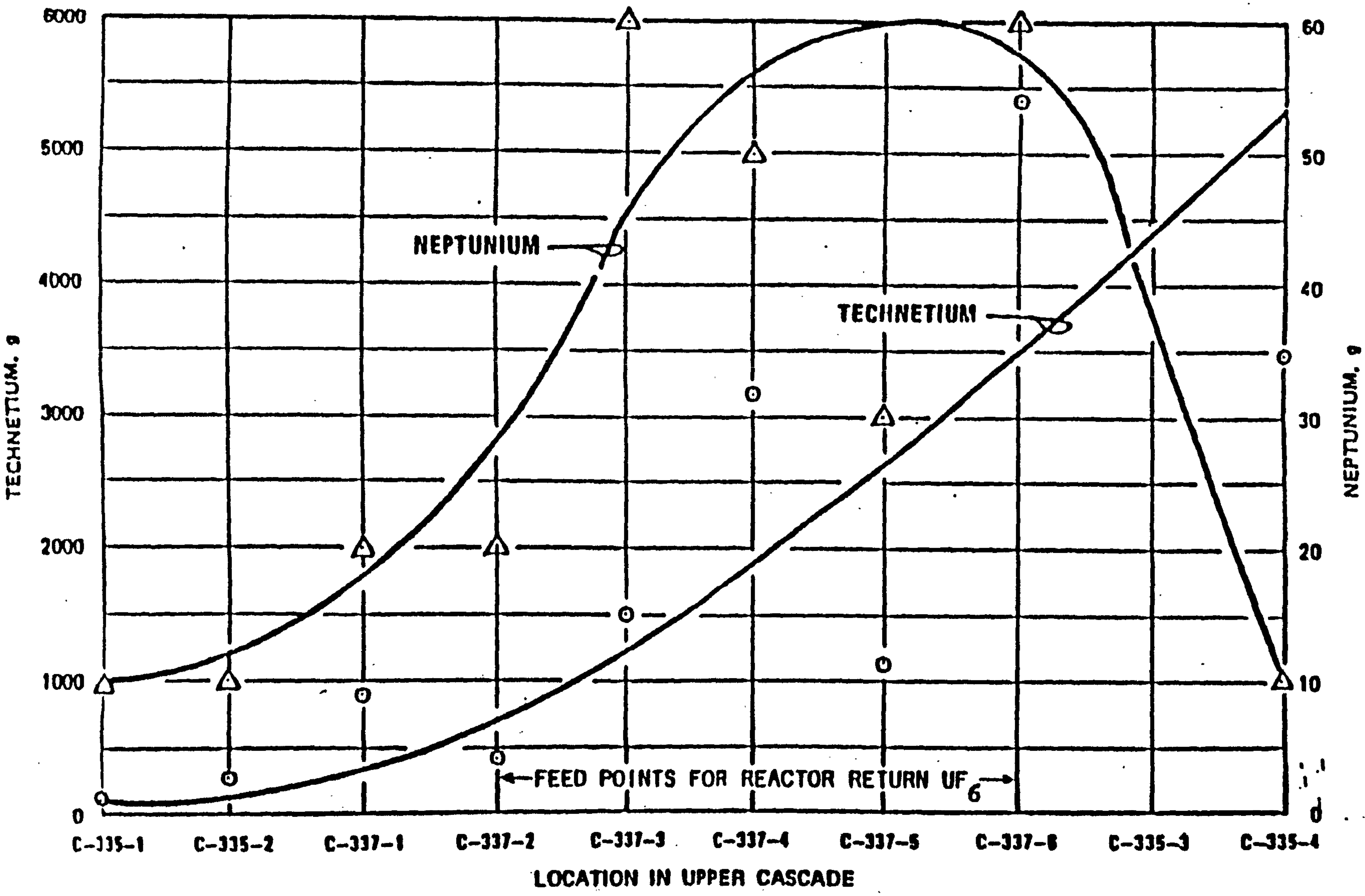
Material	Quantity Received	Quantity Fed to Cascade
Tc-99	86 kg	86 kg
Np-237	2.2 kg	0.54 kg
Pu-239	44 g	0.065 g

The Paducah Plant has reported that the plutonium was immobilized in the immediate areas of the feed points, and that only the neptunium exhibited any mobility at all. The next slide shows the estimated distribution of neptunium in the Paducah cascade.

SLIDE 6

This is based on actual sampling of materials from the cascade equipment. Although the data are represented by smooth curve with a single maximum, it has been pointed out that the data might have indicated a double peak distribution around the feed points. In any event, most of the Np is in the region of the cascade bracketed by the feed points.

If we assume a similar distribution for this plant the peak distribution would fall in the portion of the cascade involved in the most recent nprovement program. In this program the cells with the highest interstage lows were targeted for barrier uprating, and these were the cells at and close to the feed points. In some of the converters removed from this region the barrier was of sufficiently high quality that the converters were used to



DEPOSITION OF NEPTUNIUM AND TECHNETIUM IN THE UPPER CASCADE OF PADUCAH replace some of the stages in regions of lower flow rates where barrier quality was not so critical. Time has not permitted a close tracking of these individual cells to date, but it is clear that most of the neptunium would been removed and would have entered the waste stream from the equipment decontamination activities.

This might represent reducing the curve to zero over most of its range, say from C-335-2 at the low end to C-335-3 at the upper end. Obviously if we want to apply the Paducah experience to ORGDP it will be necessary to determine which cells at ORGDP correspond with which cells at Paducah and to determine the regions of the neptunium curve which were impacted. The tracking of converters removed from the central region and placed in other parts of the cascade will also be needed to complete the picture.

About 621 MTU of reactor returns have been fed at ORGDP since 1974. Of the transuranics and fission products contained in these 621 MTU, the presumption is perhaps reasonable that, except for Tc, the impurities from 420 MTU were removed during the Cascade Improvement Program. Some fraction of the transuranic and fission products in the remaining 200 MTU would also have been removed (post-1980); therefore, the cascade currently contains these classes of impurities from less than 200 MTU of reactor returns and could be considered relatively clean with respect to these materials.

Small quantities of French power reactor returns (Cogema) were also fed at ORGDP. This material is very clean as illustrated in the following typical analysis.

SLIDE 7

TYPICAL COGEMA REACTOR RETURN ANALYSIS

Item	Specification	Analysis
Fission Product Gamma (% of aged natural uranium)	20	6
Fission Product Beta (% of aged natural uranium)	10	0.1
Transuranic Alpha	1500	8.13

We may conclude that the small quantities of French reactor returns fed have had no significant effect on the amounts of fission products or -ansuranic elements in the ORGDP cascade.

Health Physics Aspects of Plutonium and Neptunium in the Cascade

At the time equipment was being removed from the cascade during the past Cascade Improvement Program the concentrations of neptunium and plutonium in the plant equipment were probably at the highest levels in the history of the plant. While this program was in progress an intensive air monitoring program was instituted by ORGDP Health Physics to evaluate the exposure potentials associated with neptunium, plutonium, technetium, and thorium-230 in the cascade equipment. Thorium-230 is the daughter product of uranium-234 and the amount of this isotope in the plant would be increasing as time passed. The key results of this program are shown on the next slide. Results are based on 127 samples, approximately 50% breathing zone and 50% general air in the immediate vicinity of the operation being sampled.

SLIDE 8

AIRBORNE CONCENTRATIONS OF ISOTOPES LISTED

	Radiation Protection Guide* dpm/m^3			
Isotope	DOE 5480.1A Chapter 11	DOE 5480.11	Maximum dpm/m^3	Average dpm/m^3
Np-237	8.88	4.44	4.6	0.11
Pu-239	4.44	4.44	0.8	0.04
Th-230	4.44	6.66	0.97	0.04

*Lowest of the values listed.

The results of these measurements indicate that the concentrations of Pu, Np, and Th-230 are well within the range where safety measures prescribed for the handling of uranium materials will also provide adequate protection from the associated transuranics and thorium-230. Thus, no special precautions will be required for equipment maintenance or removal associated with desensitization or D&D operations.